28-29 September 2023,
Kragujevac, Serbia

www.iccbikg2023.kg.ac.rs

2nd International Conference on Chemo and Bioinformatics
ICCBIKG_2023

ICCBIKG
International Conference on Chemo and BioInformatics

BOOK OF PROCEEDINGS

2nd International Conference on Chemo and BioInformatics

ICCBIKG 2023

BOOK OF PROCEEDINGS

September 28-29, 2023

Kragujevac, Serbia

Sponsored by

2nd International Conference on Chemo and BioInformatics, Kragujevac, September 28-29, 2023, Serbia.

Editors:

Professor Dr. Zoran Marković

Professor Dr. Nenad Filipović

Technical Editors:

Jelena Živković

Marko Antonijević

Dr. Žiko Milanović

Dr. Vladimir Simić

Proofreading:

Marijana Dimović

Publisher:

Institute for Information Technologies, University of Kragujevac, Serbia, Jovana Cvijića bb, 2023

Press:

„Grafo Ink“, Kragujevac

Impression:

120 copies

CIP - Каталогизација у публикацији - Народна библиотека Србије, Београд

54:004(048)(0.034.2)
57+61]:004(082)(0.034.2)

INTERNATIONAL Conference on Chemo and BioInformatics (2 ; 2023 ;
Kragujevac) Book of Proceedings [Elektronski izvor] / 2nd International Conference on
Chemo and BioInformatics, ICCBIKG 2023, September 28-29, 2023 Kragujevac,
Serbia ; [editors Zoran Marković, Nenad Filipović]. - Kragujevac : University,
Institute for Information Technologies, 2023 (Kragujevac : Grafo Ink). - 1 USB
fleš memorija ; 1 x 2 x 6 cm

Sistemski zahtevi: Nisu navedeni. - Nasl. sa naslovne strane dokumenta. - Tiraž
120. - Bibliografija uz svaki rad.

ISBN 978-86-82172-02-4

a) Хемија -- Информациона технологија -- Зборници b) Биомедицина --
Информациона технологија -- Зборници

COBISS.SR-ID 125908489

High-throughput screening of novel hydrogen storage materials – ML approach

K.Batalović[1*], J.Radaković[1], B.Paskaš Mamula[1], M.Medić Ilić[1], B.Kuzmanović[1]

[1] University of Belgrade, VINCA Institute of Nuclear Sciences – National institute of the Republic of Serbia, Department of Nuclear and Plasma Physics, CONVINCE - Center of Excellence for Hydrogen and Renewable Energy, Mike Petrovića Alasa 12-14, Belgrade, Serbia; e-mail: kciric@vin.bg.ac.rs

* *Corresponding author*

DOI: 10.46793/ICCBI23.580B

Abstract: Hydride formation in metals is a widely studied and applied phenomenon necessary to transition to clean energy solutions and various technological applications. We focus on three perspective applications of these materials, namely near-ambient hydrogen storage, hydrogen storage compressor materials, and alkali metal conversion electrodes, to demonstrate acceleration in the research achieved by utilizing a data-driven approach. Graph neural network was developed using a transfer learning approach from the MEGNet model and data related to the thermodynamics of hydride formation obtained in experimental work. Based on the crystal structure and composition as input features, we apply the MetalHydrideEnth model developed in our previous work to predict hydride formation enthalpy in intermetallic compounds. In this work, we focus on demonstrating how this approach, combined with available crystal information obtained from density functional theory calculations, can be applied for fast and extensive searches of novel metal hydride materials, having in mind the above-listed applications.

Keywords: metal hydride, GNN, hydrogen storage, DFT, conversion-type anode

1. Introduction

The storage of hydrogen, a clean energy vector, is essential in adopting various renewable energy use cases. Storing hydrogen by chemical bounding in the metals is a widely studied and applied method that offers safe and efficient storage [1]. In addition, specific applications, including metal hydride-based compressors and alkali metal hydride electrodes, also depend on tuning the stability of metal hydrides [2]. Metal hydrides form reversibly during hydrogen sorption in the metal/intermetallic compound. The enthalpy change (expressed in kJ/molH_2) in the hydride formation reaction determines the metal hydride's applicability [1].

Data-driven science as the 4th paradigm in materials science [3] has led to the rapid development of materials informatics, and dedicated machine learning approaches for materials design [4]. Although several exciting machine learning models were developed to predict the capacity and thermodynamical features of the metal hydride

materials, in this work, we focus on the model based on the graph neural network implementation for the crystal materials, MEGNet, which demonstrated low prediction errors in a broad array of properties in both molecules and crystals, enabling hydride formation energy prediction with DFT accuracy [5]. Using a transfer learning approach and a combination of DFT crystal structures and experimentally measured enthalpies of hydride formation, the MetalHydrideEnth [6] model was developed in our recent work [7]. We perform a large-scale screen of a Material Project database [8] for possible metal alloy compositions, predict the enthalpy of hydride formation, and discuss the findings considering these materials' applications.

,

2. Methodology

Elemental embeddings are adopted from the mp-2019.4.1 model [15] (trained on a data set containing 133,420 data from the Materials Project (MP) [8]). The model comprises 2 MEGNet blocks (each with two dense layers with 12 and 8 nodes per attribute) trained on the dataset containing experimental heats of forming various metal hydrides paired with the composition and crystal structure of the corresponding metal/intermetallic compound [7]. In particular, the structural features and chemical composition of metal/intermetallic compounds are transformed into an index-type graph and used to predict the heat of hydride formation. The model showed an MAE of 9.1 kJ/molH_2 for predicting hydride formation enthalpy of arbitrary intermetallics [7]. We used the Materials Project database [8] to provide the composition and crystal structure of the metal/intermetallic compound; the database was queried using pymatgen API [9].

3. Results

3.1 Near ambient hydrogen storage and conversion type anodes for Li-ion batteries

For hydrides to reach an equilibrium pressure of 1 bar at 300K, classifying them as suitable for near-ambient hydrogen storage, an enthalpy change of -39.2kJ/molH_2 in the hydride formation reaction is required [1]. In addition, high stability of the material and friendly use are desired to maintain multiple sorption cycles. A low intermetallic mass is a starting point for obtaining a high gravimetric density of stored hydrogen. The highest stability of the proposed intermetallics was requested by limiting the search to intermetallics having formation energy per atom at the convex hull of thermodynamics. The second condition is met by adopting restrictions in terms of compositions: we excluded alloys containing exotic, radioactive, or highly toxic elements, and for these applications, we focus on the Mg-containing intermetallics. This narrowed the metal alloy compositions from over 19000 intermetallics containing Mg to 433 stable intermetallics satisfying the above conditions. Fig.1 displays the predicted enthalpy of hydride formation in these intermetallics as a function of the stability of the alloy itself. Thirty-two potential candidates [10] are identified as suitable for near ambiental storage (fig.1., orange shaded). At the same time, the stability of corresponding metal alloys,

shown as formation energy per atom of the alloy, is found in the range -0.8eV to 0eV.

We examine the same batch of materials for conversion-type anode materials in Li-ion batteries. MgH_2 conversion electrodes improve capacity and density while alloying Mg improves durability and enables the selection of suitable working potentials.

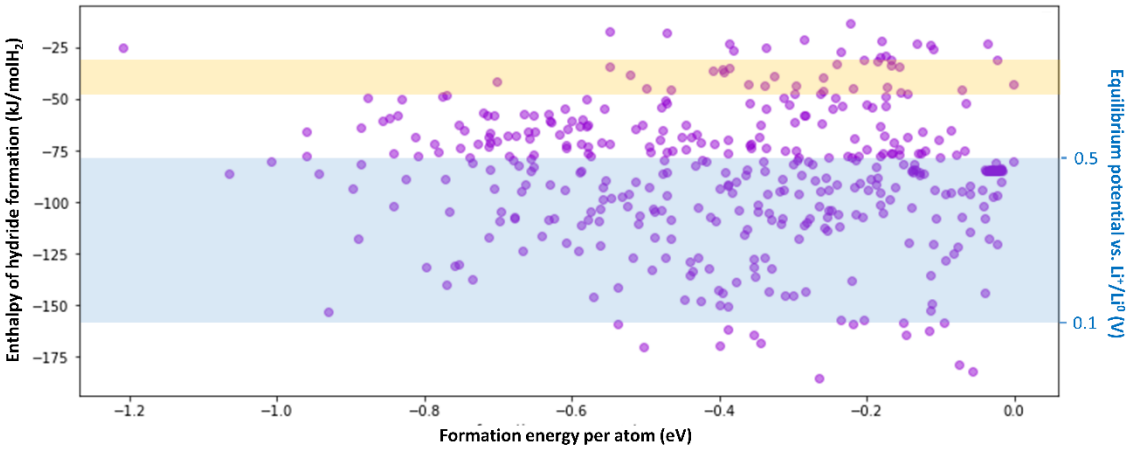

Figure 1. Fig.4. Predicted enthalpy of hydride formation and equilibrium potential vs. Li^+/Li^0 as a function of formation energy per atom in the intermetallic compound.

For negative electrodes in Li-ion batteries, the safe potential range is 0.1–0.5 V versus Li^+/Li^0. Potential vs. Li^+/Li^0 is calculated from the known changes in Gibbs free energy in the formation of LiH and metal hydride [11]. A wide range of potentials related to the Mg-containing intermetallics can be obtained (Fig. 1). Hydrides of the same or higher stability as compared to MgH_2 are needed, and a total of 244 potential candidates [10] are selected for this application (shaded blue, fig.1.).

3.2 Metal hydride compressor materials

An additional search is done for high-pressure hydrogen output materials. In particular, AB_2 intermetallics (A=Ti, Zr and B= Mn, Cr, Co) are assessed for utilization in metal compressors. To be used for this application, the lower stability of hydride is required as compared to storage materials. Table 1 lists predicted enthalpies of hydride formation in these materials, enabling the structural and compositional tuning of hydride stability.

Table 1. Predicted enthalpies of hydride formation for high-pressure hydrogen output materials

	ΔH ($kJ/molH_2$)	
A=	Ti	Zr
ACr_2	-19.9 *Laves phase* -19 *cubic*	-41.0 *Laves phase* -39.4 *cubic*
AMnCr	-18.3	-35.3
ACo_2	-15.8 *Laves phase* -14.1 *cubic*	-26.2 *cubic*

3. Conclusions

We demonstrate the application of a dedicated machine learning model for fast screening of intermetallic compounds based on the stability of their hydrides. Based on the structural features and composition of intermetallic compounds we are able to predict their behavior in 3 various systems – near ambient hydrogen storage, high-pressure hydrogen output materials, and conversion-type electrodes in Li-ion batteries.

Acknowledgment

This research is funded by the Ministry of Education and Ministry of Science, Technological Development and Innovation, Republic of Serbia (contract number: 451-03-47/2023-01/200017).

References

[1] V. Yartys, M. Zhu, *Recent advances in hydrogen storage materials,* J. Alloys Compds., 927 (2022) 166892.
[2] A. El Kharbachi, E. M. Dematteis, K. Shinzato, S. C. Stevenson, L. J. Bannenberg, M. Heere, C. Zlotea, P. Á. Szilágyi, J.-P. Bonnet, W. Grochala, D. H. Gregory, T. Ichikawa, M. Baricco, B. C. Hauback, *Metal Hydrides and Related Materials. Energy Carriers for Novel Hydrogen and Electrochemical Storage*, J. Phys. Chem. C, 124 (2020) 7599–7607.
[3] G.R. Schleder, A.C.M.Padilha, C.M.Acosta, M.Costa, A.Fazzio, *From DFT to machine learning: recent approaches to materials science–a review*, J. Phys.: Mater., 2 (2019) 032001.
[4] C.Chen,Y.Zuo,W. Ye,X.Li,Z.Deng,S. Ping Ong, *A Critical Review of Machine Learning of Energy Materials*, Adv. Energy Mater., 10 (2020) 1903242.
[5] C. Chen, W. Ye, Y. Zuo, C. Zheng, S. Ping Ong, *Graph Networks as a Universal Machine Learning Framework for Molecules and Crystals*, Chem. Mater, 31 (2019) 3564–3572.
[6] K. Batalović, J. Radaković, B. Kuzmanović, M. Medić Ilić, B. Paskaš Mamula, *"MetalHydrideEnth"*, Mendeley Data, V1 (2022), doi: 10.17632/4tpmdzxtf6.1.
[7] K. Batalović, J. Radaković, B. Paskaš Mamula, B. Kuzmanović, M. Medić Ilić, *Predicting the Heat of Hydride Formation by Graph Neural Network - Exploring the Structure–Property Relation for Metal Hydrides*, Adv. Theory Simul., 5 (2022) 2200293.
[8] A. Jain, S.P. Ong, G. Hautier, W. Chen, W. Davidson Richards, S. Dacek, S. Cholia, D. Gunter, D. Skinner, G. Ceder, K. A. Persson, *Commentary: The Materials Project: A materials genome approach to accelerating materials innovation*, APL Materials, 1 (2013) 011002.
[9] 29. S. Ping Ong, W. D. Richards, A. Jain, G. Hautier, M. Kocher, S. Cholia, D. Gunter, V. Chevrier, K. A. Persson, G. Ceder, *Python Materials Genomics (pymatgen) : A Robust, Open-Source Python Library for Materials Analysis*, Comput. Mater. Sci., 68 (2013) 314–319.
[10] K. Batalović, J. Radaković, B. Paskaš Mamula, B. Kuzmanović, M. Medić Ilić, *Machine learning-based high-throughput screening of Mg-containing alloys for hydrogen storage and energy conversion applications*, J. Energy Storage, 68 (2023) 107720.
[11] Y. Oumellal, A. Rougier, G. A. Nazri, J-M. Tarascon, L. Aymard, *Metal hydrides for lithium-ion batteries*, Nat. Mater., 7, 916-921, 2008.